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Spinodal vaporization – a critical and imposing decay mode of highly excited nuclear systems

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ABSTRACT

A novel prompt decay mode of highly excited nuclear systems is shown to set in with necessity as a certain critical excitation energy per nucleon is reached. It is driven by a peculiar, open-ended type of spinodal instability, unique to self-bound open systems, and consists in parts of the system undergoing spontaneous indefinite thermal expansion ending in vaporization into the surrounding open space. The mode, named here spinodal vaporization, is distinctly different from all known decay modes of excited nuclei and faces no competition from the latter. It sets a natural upper limit for the excitation energy that can be thermalized by compound nuclear systems, while setting also a limit to the applicability of thermodynamics to the description of highly excited nuclear systems.

1. Introduction

Understanding the limits of thermodynamical stability of excited nuclear systems has been a focus of numerous theoretical and experimental studies [1–5] from the dawn of nuclear science. Theoretical studies have so far been overwhelmingly concentrated on instabilities in bulk nuclear matter kept under controlled conditions and, notably, at a fixed volume allowing under certain circumstances for an unphysical buildup of a gas of free nucleons in thermodynamical equilibrium with the liquid phase. The model calculations show that under such condition, owing to the Van der Waals-like equation of state for uniform matter, three kinds of domains exist in the space of controlling variables for such matter. Firstly, there are domains where uniform nuclear systems would be stable. Then, there are domains where the uniform matter would be metastable and where, given time, it would end up as a two-phase system as a result of evaporation or condensation. And, finally, there is a domain commonly named spinodal, where uniform matter would be unstable and would spontaneously separate into liquid and gaseous phases. The above domains are readily identifiable, e.g., on the standard plots of Van der Waalsian isotherms as functions of specific volume and pressure.

In contrast to the above theoretical view, experimental studies necessarily involve finite nuclear systems formed in the course of nuclear reactions. Such systems are not subject to external confinement and are thus at best metastable – free to evaporate particles into open space and to undergo shape fluctuations leading to fragmentation – the two basic statistical decay modes of metastable excited compound nuclei. It is worth noting here that this metastability is at the very crux of sound thermodynamical models of nuclear decay, such as Weisskopf's evaporation model [6] and various models of compound-nuclear fission, as well as various numerical implementations of these models [7–10]. The lack of external confinement has, however, one other unavoidable consequence which is thermal expansion. The paramount importance of the latter at elevated excitation energies has been largely overlooked in theoretical modeling of the equilibrium-statistical decay of excited nuclear systems – other than those reported in precursor conference proceedings [11] of the present study.

The present work is part of a continued effort [11–22] to construct an open microcanonical framework for understanding decay phenomena of highly excited metastable nuclear systems produced in the course of heavy-ion collisions and the limits of compound-nuclear metastability of such systems. It reveals the crucial and hitherto unappreciated role of thermal expansion in a peculiar *thermal* spinodal destabilization of highly excited nuclear systems. The latter gives rise to a hitherto unknown prompt decay mode of such systems, named here spinodal vaporization, which then imposes a natural upper limit on excitation energy, a compound-nuclear system is capable of thermalizing and a limit on the domain of validity of the very concept of a compound nucleus. It is worth noting that while the existence of a limit on such validity

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has always been understood, beginning with the seminal work by Weisskopf, [6] its actual location on the excitation energy scale has not been accurately specified.

The term "open microcanonical" is used in this study to stress the fact that, unlike conventional microcanonical models of statistical nuclear decay, the formalism used here explicitly acknowledges lack of external confinement and thus allows for both, large-scale shape fluctuations and thermal expansion. Note, that the lack of external confinement translates into the pressure at the periphery of the system (microcanonical pressure) being zero regardless of the excitation energy or (microcanonical) temperature, which clearly is the case for excited atomic nuclei residing in vacuum.

2. Outline of the theoretical framework

The present study utilizes elements of a thermodynamic framework developed over many years of studies of the properties of highly excited nuclear systems, both, infinite and finite [11–22]. The model is based on the fundamental thermodynamical principle that an isolated system will assume macroscopic configurations fluctuating around the one that shows highest Boltzmann entropy, whenever such exists. These maximum-entropy configurations are homogeneous for infinite systems and isotropic with a smooth density profile, for finite systems. As infinite systems one generally understands systems that are large enough to warrant neglect of the inhomogeneous surface domain. In contrast, the latter surface domain is of essence in modeling the behavior of finite systems. The above assumptions of uniformity and isotropicity are well justified given the nature of the formalism used. Naturally, for a self-bound object residing in vacuum, such as an excited atomic nucleus, maximum entropy is indicative of zero microcanonical pressure regardless of the excitation, as that pressure is given by the first derivative of entropy with respect to volume. For infinite systems, the pressure is then uniformly zero, while for finite system only the pressure at the periphery (microcanonical pressure) is zero, while in the interior bulk matter it is greater than zero as a result of the action of the surface tension. In the latter systems, the surface tension decreases with increasing excitation energy and so does the pressure of the bulk matter.

Boltzmann entropy is evaluated in the framework of zero-temperature Fermi gas model and Thomas Fermi approximation. The zero-temperature formalism was utilized for the sake of simplicity, but it is also a reasonably good approximation in the range of excitation energies considered. Thus, Boltzmann entropy is expressed as:

$$S_{config} = 2\sqrt{a_{config}(E - E_{config})}, \tag{1}$$

with E, a_{config}, and E_{config} denoting the total energy, the level density parameter and the interaction energy of the configuration considered, respectively. The latter represents the zero-temperature energy of the configuration considered and includes potential energy and the Pauli kinetic energy of the Fermi matter. Eq. (1) is the base equation of the model, allowing one to evaluate S_{config} for any configuration of interest characterized solely by the matter density distribution $\rho_{config}(\vec{r})$.

The level density parameter a_{config} was calculated using the formalism proposed in Ref. [12]:

$$a_{config} = \alpha_o \rho_o^{2/3} \int \rho(\vec{r})^{1/3} d\vec{r}, \tag{2}$$

where α_o expresses the value of the level density parameter per nucleon at normal matter density ρ_o.

The interaction energy is here calculated by folding a standard Skyrme-type EOS interaction energy density $\epsilon_{int}^{EOS}(\rho)$ with a Gaussian folding function and the folding length, adjusted so as to approximately reproduce the experimental surface diffuseness of finite droplets of nuclear matter.

$$E_{int}^{EOS} = R_{Gauss} \int \epsilon_{int}^{EOS}\left(\rho(\vec{r} - \vec{r}')\right) e^{-\frac{(\vec{r}-\vec{r}')^2}{2\lambda^2}} d\vec{r} d\vec{r}'. \tag{3}$$

For the equation of state, the present study adopted a standard form consistent with Skyrme-type nucleon–nucleon interaction, which implies the interaction energy density (appearing in Eq. (3)) in the form of

$$\epsilon_{int}^{EOS}(\rho) = \rho\left[a\left(\frac{\rho}{\rho_o}\right) + \frac{b}{\sigma + 1}\left(\frac{\rho}{\rho_o}\right)^{\sigma}\right] \tag{4}$$

The values of the parameters a, b and σ in Eq. (4) are determined by the requirements for the binding energy, matter density, and the incompressibility modulus to have prescribed values. The values chosen in this study of $a = -62.43$ MeV, $b = 70.75$ MeV, and $\sigma = 2.0$ imply a normal density of $\rho_o = 0.168$ fm^{-3}, binding energy per nucleon at normal density of $\epsilon_{EOS}/\rho_o = -16$ MeV, the incompressibility modulus of $K = 220$ MeV, and Fermi energy at normal density of $E_0^{Fermi} = 38.11$ MeV.

One notes that in the model calculations for uniformly distributed matter, the finite range of interaction is of no consequence and the configuration energy can be written simply as $E_{config} = V\epsilon_{EOS}(\rho)$, where V is the system volume. One notes also, that the results of the calculations do not depend critically on the actual parameterization of the equation of state and that a simple harmonic interaction results in similar overall trends as the one used in the present study.

At the crux of the present study lies a particular kind of spinodal instability unique to open microcanonical systems. In this respect, it is worth recalling that spinodal instabilities are always associated with certain extensive state variables of the system being pairwise fixed, with ratios of these extensive variables then representing derived intensive variables, such as the matter density ρ, excitation energy per nucleon E/N, and iso-asymmetry per nucleon $(N - Z)/A^2$. The important difference between the "true" and derived intensive variables is that the former (such as temperature T, pressure p, chemical potentials for protons and neutrons μ_p and μ_n) are idealizations and are formally fixed throughout the system by *fiat* of the ensemble considered, while the latter are being fixed only on average, allowing for fluctuations of their values across the system up to allowing for coexistence of liquid and gaseous phases with different matter densities, excitation energies per nucleon, and iso-asymmetries per nucleon. The importance of the derived intensive variables stems from the fact that the equation of state refers to their local values and so do the characteristic state functions for uniform matter, proper for ensembles considered. For the matter to be stable for a particular kind of thermodynamic ensemble, the characteristic state function for this ensemble must be either a convex (Helmholtz free energy for canonical ensemble and Gibbs free energy for isothermal-isobaric ensemble for iso-asymmetric systems) or a concave (entropy for microcanonical ensemble) function of its derived intensive variables. In mathematical terms, this means that Hessian (curvature matrix) of this characteristic state function must be either positive definite or negative definite, respectively.

In view of the above, it is clear that the character of possible spinodal instability not only depends critically on the kind of thermodynamical ensemble considered, but is also unique to this kind, as different ensembles fix different sets of state variables. And so, in the classical canonical ensemble, fixed are (intensive variable) temperature T, and extensive variables of volume V and the number of particles N, allowing under circumstances for appearance of concavity of the Helmholtz free energy with respect to matter density and the resulting purely *mechanical* spinodal instability and

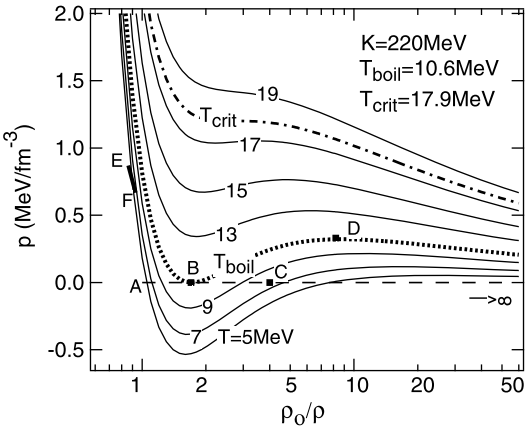

Fig. 1. Isotherms for the model matter. The isotherm corresponding to zero-pressure boiling temperature is shown in dotted line and the critical isotherm is shown in dash-dotted line. The adiabatic trajectory for a hypothetical infinite system at zero pressure is shown in dashes (line AC), while such for the bulk of a finite (A = 100) system is shown in bold solid line (line EF, see text). Point B represents both, the lower (low specific volume) limit of the *mechanical* (canonical ensemble) and the lower (low excitation energy) limit of the *thermal* (open microcanonical ensemble at zero pressure) spinodal domains, while points D and C represent the upper (high specific volume or high excitation energy) limits of these domains, respectively.

ultimately liquid-gas coexistence. In contrast, in the classical microcanonical ensemble fixed are the extensive variables V, N and the total energy E, allowing this time under circumstances for convexity of entropy as a function of two derived intensive variables, ρ and E/N. The corresponding Hessian is here a non-diagonal 2 by 2 matrix, with eigenvectors not aligned with either coordinate axis. Thus the possible spinodal instability will be of mixed *thermo-mechanical* character and will also ultimately result in coexistence of liquid and gaseous phases with different densities and excitation energies per nucleon. For the open microcanonical ensemble considered in the present study, entropy is a function of the one derived intensive variable of E/N only, and its Hessian degenerates into a single-element (1 by 1) matrix. Accordingly, the possible spinodal instability here is of purely *thermal* character. In this case, the instability does not, however, lead to phase coexistence even as it leads first to phase separation. This is so because the gaseous phase is not self-bound and will inevitably vaporize further into the surrounding vacuum, synonymous with a prompt spontaneous decay of the system considered.

For the sake of completeness it is worth noting that spinodal instabilities are absent altogether in ensembles which are defined in terms of only one extensive state variable in addition to the requisite number of intensive variables. Such ensembles do not fix (on the average) any derived intensive variables. They include the grand canonical ensemble, with the sole extensive variable being volume V and intensive variables being chemical potential μ and temperature T, and the isothermal-isobaric ensemble for isoneutral nuclear matter with the sole extensive variable being this time the number of particles, while intensive variables being temperature T and pressure p.

3. Results

The general behavior of uniform Fermi matter can be well understood from the appearance of isotherms in the familiar Van der Waals type plots. These are illustrated in Fig. 1 for the bulk model matter with Skyrme-type EOS with a compressibility constant of K = 220 MeV. The isotherms were constructed using microcanonical approach, with T and p representing microcanonical temperature and pressure, and are identical to those computed in a canonical approach, in virtue of ensemble equivalence for uniform matter. The isotherms feature prominently the familiar *mechanical* spinodal domains of negative compressibility characteristic for canonical ensemble (such as section BD of the dotted isotherm in Fig. 1) at constant temperature T and volume V and such ensemble only. Note, however, that for nuclear systems, canonical ensembles are of purely academic interest only, as they cannot be implemented practically. Importantly, the isotherms in Fig. 1 feature also, albeit less conspicuously, the crucial *thermal* spinodal domain of negative heat capacity for a realistic self-bound open microcanonical ensemble in vacuum, i.e., at zero pressure. This domain is bounded at point B on one side and point C on the other. Point B is by construction the one where the zero-pressure isobar is tangential to a particular isotherm at the minimum of the latter, the isotherm being the one defining the boiling-point temperature. In a canonical representation, this isotherm has been termed in recent past as "flashing point" isotherm. [23] Point C represents the excitation energy at which the system becomes unstable against uniform indefinite expansion and in that sense the spinodal domain BC is here open-ended. In this domain BC nuclear matter will undergo phase separation, with part of it condensing toward point B and the other part vaporizing toward point C and beyond, i.e., undergoing vaporization.

That the domain BC in Fig. 1 is indeed *thermal* spinodal is evident when one follows the evolution of a self-bound system at zero pressure (isobaric system) with increasing excitation energy per nucleon, starting at point A in Fig. 1 (normal density at zero excitation and zero external pressure) towards point B, and beyond. The excitation is measured here (for the correspondence between the excitation energy and specific volume see Fig. 2) by the specific volume displayed on the abscissa and the trajectory of such an *isobaric at zero pressure* evolution is illustrated by the dashed line AC. One notes readily, that first, the temperature rises with increasing specific excitation energy, while the system undergoes thermal expansion. This rise in temperature is indicated by crossing of isotherms with progressively higher temperature indices as one proceeds from point A to point B. However, beginning at point B, the trend reverses and now isotherms are being crossed with progressively lower temperature indices, indicative of the heat capacity becoming formally negative. This indicates that at point B the system enters (*thermal*) spinodal domain of convexity of Boltzmann entropy as a function of excitation energy and, thus becomes unstable against phase separation. Not only can it not exist in this domain as uniform matter, neither can it exist here as a two-phase system. This is so, because the vapor phase is not self-bound and will expand indefinitely at the expense of its own energy content upon reaching spinodally (at the expense of energy derived from neighboring parts) point C in Fig. 1. Which means, that upon acquiring excitation energy in excess of what is needed to reach the "boiling" point B, the system will promptly shed a portion of itself via a boiling-like process named here spinodal vaporization, leaving behind a "leaner" metastable residue at point B. The latter will subsequently decay statistically via well-known decay modes of evaporation and fragmentation showing the source temperature as that at point B, regardless of the initially acquired excitation energy per nucleon. One may posit that the spinodal vaporization is akin to boiling, even as in practical realizations the latter may to some extent rely on nucleation phenomena. The isotherm labeled as T_{boil} in Fig. 1 is by definition the one that is tangential to the zero-pressure line in its minimum.

The purpose of Fig. 1 is to demonstrate that any self-bound Van der Waalsian system (one that features characteristic Van der Waalsian isotherms) at zero pressure would enter the spinodal domain with necessity, and not by *fiat* of modeling, as soon as its initial energy is raised in excess of that at point B, and that the

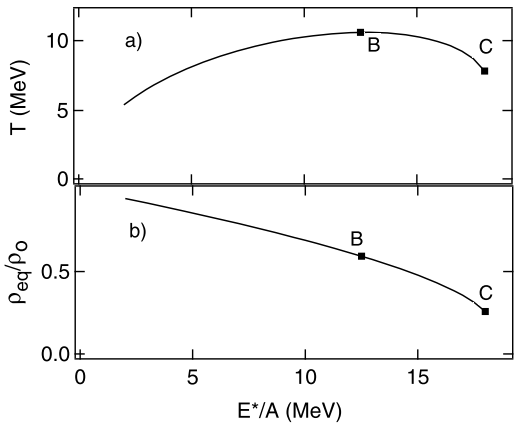

Fig. 2. Caloric curve (panel a) and equilibrium density (panel b) as functions of excitation energy for a self-bound Fermi liquid under zero pressure. Points B and C correspond to those shown in Fig. 1.

vaporization must take place regardless of whether any "conventional" statistical decay channels are open or not.

While Fig. 1 proves the inevitability of spinodal vaporization or boiling for any Van der Waals – like matter at elevated excitations and demonstrates that the phenomenon in question has been in "plain sight" for all these years, a more conventional depiction of the instability is offered in Fig. 2 in the form of a caloric curve for the open-microcanonical system considered. As seen in this figure, the caloric curve for such a system features prominently a domain of formal negative heat capacity, i.e., *thermal* spinodal instability. This domain ends formally at point C where the system becomes unstable with respect to uniform expansion, such that any portion of vapor reaching (spinodally) that point would expand further indefinitely. It is worth noting that the matter density behaves here monotonically with increasing excitation energy in the domain of interest, even as thermal expansion is here at the crux of the spinodal instability. As pointed out earlier in Section 2, *thermal* spinodal instability is unique to open iso-neutral microcanonical systems, the only ones where the Boltzmann entropy depends solely on total energy per nucleon and the only ones where the convexity of entropy function translates directly into the formally negative heat capacity. In contrast, for iso-asymmetric systems spinodal instabilities in open microcanonical ensembles would be of mixed *thermo-chemical* character [11].

The onset of spinodal vaporization is further illustrated in Fig. 3 for a system of two equal masses in thermal contact, in a purely open microcanonical picture that allows for thermal expansion while keeping constant the total excitation energy and mass number, but not the volume. As seen in this figure, at low and moderate excitation energies, the configuration of maximum entropy is that where both equal parts have equal excitation energies, indicating uniform matter and excitation energy distribution. In this regime, the system may and will continually fluctuate away from uniformity, but the negative feedback of concave entropy will return the system back toward uniformity. The situation is dramatically different at excitation energies past the boiling point where the curvature of the entropy surface turns positive. From this point on, any acquired asymmetry would be further reinforced by the action of entropy driving now the system further away from uniformity, with one portion eventually vaporizing away into the surrounding space. It is worth noting that in the spinodal domain of excitation energy, there is no hope of ever seeing the system at a state of approximate uniformity, even transiently.

For self-bound finite systems in vacuum, i.e., at zero external pressure, the presence of the surface domain alters the character of the possible spinodal instability in an important way. Here, the

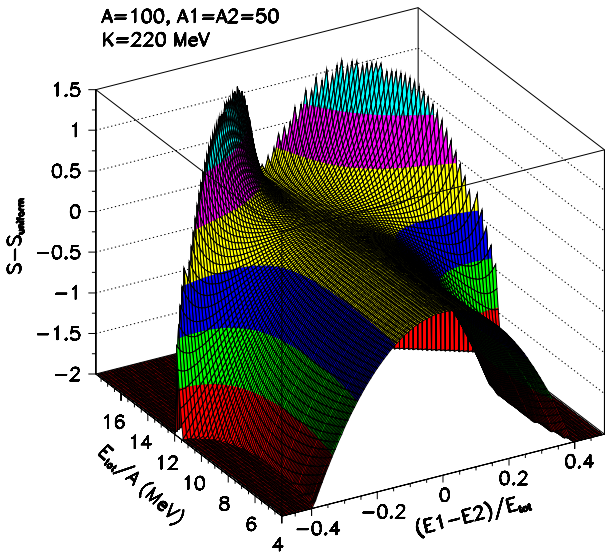

Fig. 3. (Color online.) Reduced two-phase configuration entropy surface for a configuration of two equal-A subsystems with differing split of the available excitation energy E^* between these phases.

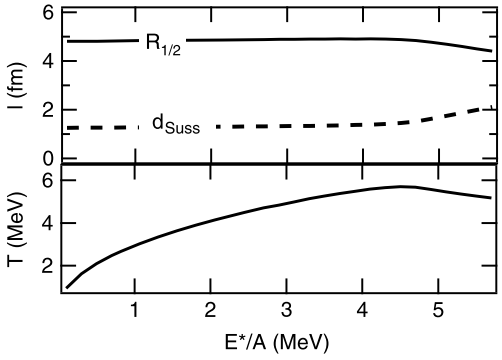

Fig. 4. Evolution of the mass density distribution parameters and the microcanonical temperature with excitation energy per nucleon (see text).

very term can no longer be associated with a matter configuration that is uniform in space, but rather with one that is solely isotropic around the center of the system and is describable in terms of a nonuniform density profile. For iso-neutral matter considered in the present study, Boltzmann entropy is still a function solely of total excitation energy, and the onset of spinodal instability can be still inferred from the appearance of the system caloric curve. The latter is illustrated in Fig. 4 along with the evolution of the two parameters, the half-density radius $R_{1/2}$ and the surface diffuseness (Süssmann width [24]) parameter d, that were used to parameterize the error-function like [12] matter density profile. As seen in this figure, the *thermal* spinodal instability sets now in at a significantly lower temperature than it did for bulk matter. This reflects the fact that the system has now an overall lower binding energy, but mostly the fact that the surface domain is more loosely bound than the bulk. One notes, that in the spinodal domain of a formally negative heat capacity, it is the surface domain alone that tends to expand with increasing energy, while the bulk tends actually to contract while staying at all energies safely outside of its own (*thermal*) spinodal instability domain (*vide* Fig. 2). This is indicative of yet another new type of spinodal instability, the one where sections of the system within one fraction of the solid angle increase first their surface diffuseness while harvesting energy from the neighboring sections that undergo contraction, and while cooling down in the process. Subsequently, surface domains in these

fractions vaporize into the surrounding vacuum at the expense of their own energy content, all while keeping cooling down. This process is named here surface spinodal vaporization. The trajectory of the bulk (quasi-uniform) interior matter of the finite system considered here is mapped in Fig. 1 as a segment of line labeled EF reflecting reduction of the surface tension and, thus, the pressure in the interior with increasing (microcanonical) temperature. The pressure of the bulk matter is evaluated as microcanonical pressure at the density and the specific excitation energy of the (uniform) bulk matter. Note, that the latter pressure is of academic interest only and plays no role in why and how the *thermal* spinodal instability sets in. Here, it helps understand that the *surface* spinodal instability sets already in where the bulk interior matter alone is still stable.

4. Summary

In summary, the present study reveals the importance of thermal expansion for the fate of highly excited nuclear systems. It reveals the unavoidability of eventually entering the *thermal* spinodal domain, when the excitation energy injected into the system is increased in excess of a certain critical energy per nucleon that can be naturally associated with the boiling-point temperature. The study reveals a new, hitherto overlooked, decay mode of highly excited nuclei consisting in indefinite expansion of fragments of the surface domain ending in their vaporization – a process named surface spinodal vaporization. For infinite matter (of academic interest only) the spinodal vaporization is a volume phenomenon entailing indefinite growth or vaporization of various portions of matter throughout the volume of the system. While one can freely speculate about what is really happening in the spinodal domain, the underlying thermodynamic theory is helpless in this respect. What the latter can do, however, is to tell with certainty that, when formally in spinodal domain, the system lacks metastability such that there is no local maximum in the entropy function. Hence, the system will promptly shed some parts of itself along with an unfairly large share of excitation energy via vaporization, before arriving at a state of compound-nuclear metastability. The latter is then subject to conventional nuclear thermodynamic modeling.

The present study has numerous experimental implications, with the existence of a limiting temperature being the most prominent of them. It so happens, that the experimental verification of the existence of a limiting temperature is there since very many years [2] and was, actually awaiting a plausible theoretical explanation. Further, the model calculations imply that the vaporized matter is colder than the residue, a prediction that may possibly find an experimental confirmation. They imply also an unusually low latent heat of vaporization that is measured on the excitation energy scale from the boiling point energy per nucleon to the point of the onset of instability with respect to uniform expansion – a mere few MeV per nucleon (*vide* Fig. 2). Furthermore, the nature of the model calculations is such that one can expect effects of N–Z asymmetry. Such have been, indeed, already reported elsewhere [11].

Acknowledgements

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